

AN INVESTIGATION OF THE NATURE OF THE ACTIVE
DEPOSITS OF THORIUM AND RADIUM

BY

DOROTHY BLAINE MARTIN

A. B., Southwest Missouri State Teachers College, 1940

AN ABSTRACT OF A THESIS

SUBMITTED IN PARTIAL FULFILLMENT OF THE REQUIREMENTS
FOR THE DEGREE OF DOCTOR OF PHILOSOPHY IN CHEMISTRY
IN THE GRADUATE SCHOOL OF THE
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I. INTRODUCTION

The expression "active deposit" has been used to designate the active substance which collects on any object that is exposed to a radioactive gas, either thoron, radon, or actinon. The half-life of the deposit is not that of the gas, indicating that it is composed of atoms of the successive disintegration products of the gas. The relative amounts of the constituents change with time or the degree of active disintegration.

In this investigation the active deposit has been studied by a direct method, electron microscopy, to determine the appearance of the deposit and the effect of the variation of certain factors, namely: the partial pressure of the air, the partial pressure of the radioactive gas, the presence of nuclei, and the length of exposure of a surface to the radioactive gas on the form of the deposit.

II. HISTORY

The history of the nature of the active deposit includes two phases: the research indicating the aggregation of the active deposit and the experiments, and theories concerning the origin of the aggregates. The aggregation of the active deposit into characteristic groups has been indicated by the investigations of Makower and Russ¹, Mlle. Chamié², and Harrington³. The origin of the active deposit was investigated by the same people and Jedrejowski.⁴

III. MATERIALS AND METHODS

In this investigation the technique of electron micrography was adapted to the demonstration of the nature of active deposit. This method provides both direct photographic observation of the deposit including inactive as well as active atoms and a measure of the size of the aggregates.

Three materials were used as sources of active deposit, crystalline thorium hydroxide, radon sealed in ampules, and solid radium sulfate.

IV. EXPERIMENTAL AND DISCUSSION

Electron micrographs of active deposit demonstrated that it consists of discrete particles distributed at random on the surface. Active deposits of thoron and radon were observed to contain aggregates averaging 8 mmu. and 10 mmu. respectively in diameter. From these measurements the number of atoms in the aggregates was calculated to be 1,000 to 10,000 and their weight 10^{-18} to 10^{-19} g. Thus the electron microscopic studies of the structure of active deposit by providing actual pictures of the surfaces, demonstrate directly the fact that the deposit is in a state of aggregation rather than in uniform layers.

With the new technique it was possible to make the following observations concerning active deposit:

1. In addition to the aggregates varying in composition from 1,000 to 10,000 atoms, agglomerates consisting of many aggregates have been observed.
2. No marked difference in the appearance of the active deposit is found when the substrate is changed from collodion to silica.
3. Observation of active deposit under reduced pressure (partial pressure of air 740 mm., 380 mm., 10 mm., and 10^{-3} mm.) revealed that the size of the aggregates is independent, within the experimental error, of the partial pressure of air.
4. Active deposit collected from purified radon sealed in gold ampules was found to have a film-like structure; however if a polar molecule, chloroform, is present in the collection chamber the active deposit is in a state of aggregation.
5. The effect of increasing the original quantity of radon in the collection chamber is to increase the size of the aggregates.
6. Increasing the collection time causes an increase in the size of the aggregates.

The experimental data may be discussed in regard to two general mechanisms of aggregate formation, a gas phase (colloidal mechanism), and a solid phase (surface migration mechanism). Assuming gas phase aggregation, the lack of dependence of aggregate size on the partial pressure of air, the effect of polar molecules, and the similarity of the electron-micrographs of active deposit collected on silica and collodion substrates are easily explicable. Qualitatively the increase in size of the aggregates with increasing original concentration of radon and increasing contact time seems reasonable, but a possible quantitative correlation, the proportionality of the radius of the aggregates and the two-thirds power of the quantity of disintegrated radon does not hold. It is difficult also to interpret the electron micrographs as steps in a continuous process if gas phase aggregation is considered the mechanism of formation. On the other hand by the assumption of solid phase aggregation all the experimental facts which were easily interpreted by a gas phase mechanism are equally explicable. Furthermore the variation of aggregate size with contact time and original quantity of radon appear to be in accord with the theory. The varying aspect of the electron micrographs interpreted as steps in solid phase aggregation is explicable, and also the qualitative aspect of these electron micrographs agrees with the electron micrographic examination of the formation of agglomerated metal films deposited from a molecular beam⁵ which is well established to be a surface migration phenomenon. This critical study of the experimentally determined facts and the quantitative relationships derivable from the data does not unequivocally establish either mechanism, but a surface migration mechanism appears to be favored.

V. SUMMARY AND CONCLUSIONS

1. Active deposits of thorium and radium have been established to be aggregated rather than uniform films.
2. The size of the aggregates in active deposit is independent of the partial pressure of air.
3. The presence of chloroform in the collection chamber aids the formation of aggregates.

4. The size of the aggregates increases with increasing contact time.
5. The size of the aggregates increases with increasing original concentration of the radioactive gas.
6. Neither a solid phase nor a gas phase mechanism of aggregation has been established, but the former appears to be favored.

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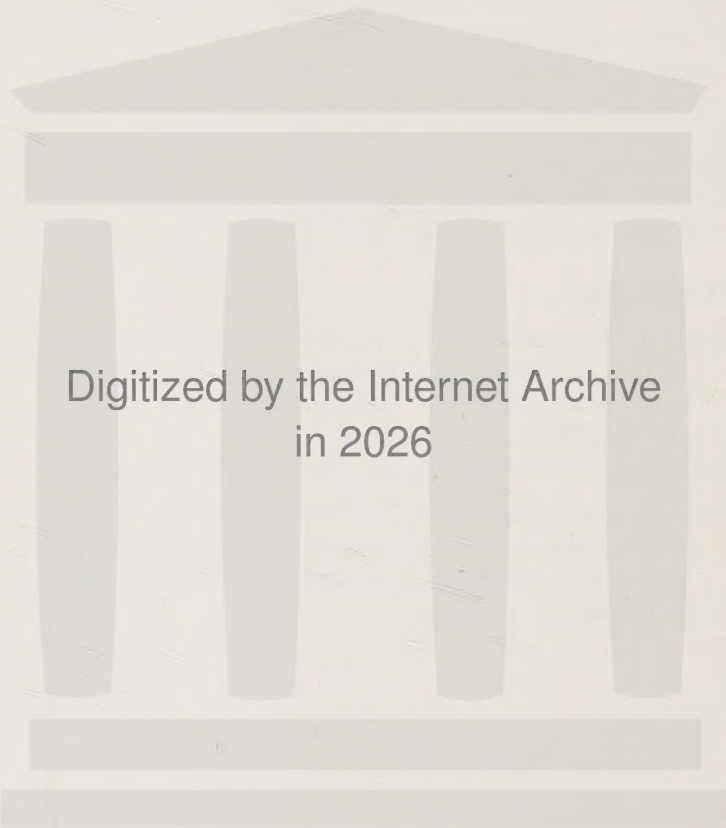
VITA

The author was born in Harwood, Missouri, September 18, 1919. She attended Greenwood training school, Springfield, Missouri, from 1924 to 1936, when she was graduated from high school. Her undergraduate work was done at Southwest Missouri State Teachers College, Springfield, Missouri, where she received the degree of Bachelor of Arts in 1940. She held assistantships at the University of Illinois during the years 1941-42, 1942-43, 1943-44, and 1944-45. She is a member of the American Chemical Society, Iota Sigma Pi, and Sigma Xi.

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